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PARTICLES IN CONVECTIVE UPDRAFTS

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NUCLEATION SCAVENGING OF SMOKE AND AEROSOL PARTICLES IN CONVECTIVE UPDRAFTS

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1. INTRODUCTION

Considerable evidence has been accumulated to suggest that nucleation scavenging may be the most important mechanism for incorporating submicron aerosol into cloud water (Radke, et al. 1980; Flossman, et al., 1985). The fraction of particles entering a cloud by nucleation scavenging depends on the maximum saturation ratio reached in the cloud. At higher saturation ratios, smaller aerosol particles become activated and are able to grow as drops, thus incorporating larger numbers of aerosol particles into the cloud drops. The maximum saturation ratio in the cloud is determined by a balance between the rate of growth of the drops and the decrease in temperature and the corresponding decrease in the saturation vapor pressure due to the ascending air parcel.

One two-dimensional cloud model has been developed which takes account of the maximum saturation ratio by self-consistently calculating cloud dynamics and the microphysical growth of drops (Hall, 1980), but three-dimensional models, which provide significantly improved representation of the dynamics of most cloud systems, are limited in their computational ability to include such effects. Thus drop size distribution and growth is not calculated in these models. Instead, the assumption is made that water condenses instantaneously if the saturation ratio becomes greater than unity. The amount of water which condenses is computed using the Clausius-Clapeyron equation along with the assumption that the saturation ratio, S_R , is always less than or equal to one.

We are testing a technique for evaluating the maximum saturation ratio that might be expected within a convective updraft whose dynamics are computed using a three dimensional cloud model. During the cloud calculation, a history of the conditions experienced by a parcel along a specific trajectory is accumulated. These "trajectory" variables are then used in a detailed microphysics model to calculate aerosol activation to drops and drop growth rates and to predict the maximum saturation ratio and the fraction of aerosol particles scavenged by nucleation to drops. The microphysics model may also be run as a stand-alone adiabatic parcel model with either fixed updraft velocities or with velocities cal-

culated self-consistently together with the other parcel variables.

In the following section, we describe our technique for coupling the microphysics model to a cloud model. The technique will be tested by comparing the results from the "trajectory" model to results obtained with a fully coupled system. In section 3, we describe some results obtained with our adiabatic parcel model which explore the dependence of saturation ratio and the scavenging of aerosols on the assumed aerosol size distribution, solubility characteristics, number concentration, and updraft velocity. Section 4 presents our conclusions.

2. COUPLING THE CLOUD MODEL AND MICROPHYSICS MODEL

We shall assume that the cloud model has been run, and a time history of the following variables along a parcel trajectory is available:

- ρ_a^t = dry air density at time t
- p^t = pressure at time t
- T^t = temperature at time t
- $\rho_{v,c}^t$ = cloud model vapor density at time t
- $\rho_{l,c}^t$ = cloud model liquid water density at time t
- $\rho_{s,c}^t$ = cloud model smoke density at time t .

The following variables are to be calculated in the microphysics model:

- $f_d(a, t)$ = number of drops with radius a at time t
- $g_{AP,d}(a, t)$ = mass of aerosol in drops of radius a at time t
- $f_{AP,u}(a, t)$ = number of unactivated aerosol particles with radius a at time t
- $g_{AP,u}(a, t)$ = mass of aerosol in unactivated aerosol particles with radius a at a time t

For an aerosol particle that contains some soluble component, the radius, a , may become larger than its dry radius, even though the aerosol has not been activated. This sub-critical growth of soluble aerosols is fully accounted for in the present model.

In order to calculate the growth rate of drops and the number of smoke particles which activate (or become cloud condensation nuclei), we need to know the degree to which the equilibrium assumption, $S_R = 1$, is violated near cloud base, i.e. we need to know how large S_R becomes. S_R will be greater than one because the updraft forces water-vapor laden air to colder regions of the cloud where it tries to condense. If this forcing occurs quickly enough, S_R will become larger than one, because the rate of condensation onto aerosol particles and drops is too slow to condense all the water that would be condensed under the equilibrium condition. Since the cloud model assumes $S_R = 1$, it is clearly not possible to maintain exact consistency between the cloud model liquid water and the microphysics model liquid water. In our attempt to couple the two models we will constrain the sum of the vapor and liquid water densities in the microphysics model to be equal to $\rho_{i,c}^t + \rho_{v,c}^t$ from the cloud model. We compute the saturation ratio and the liquid water density in the microphysics model consistent with the temperature, pressure, and dry air density determined in the cloud model. We note that use of the cloud model temperature to compute S_R introduces some error, since the effect on temperature of the difference in the amount of water which condenses in the cloud model and that which condenses in the microphysics model is not taken into account. In general, at any given time the cloud model must condense the greater amount of water. An example of the extent of the difference for a problem in which a constant updraft of 10 m/s is assumed is shown in Figure 1. The corresponding saturation ratio is shown in Figure 2. Because of this difference, the temperature predicted in the cloud model will be slightly larger than the temperature which one would calculate in a fully coupled model. Since the cloud model temperature is used to predict the saturation vapor pressure, we expect that S_R will be slightly underestimated by this method of coupling. Results will be presented at the meeting which estimate the magnitude of this error.

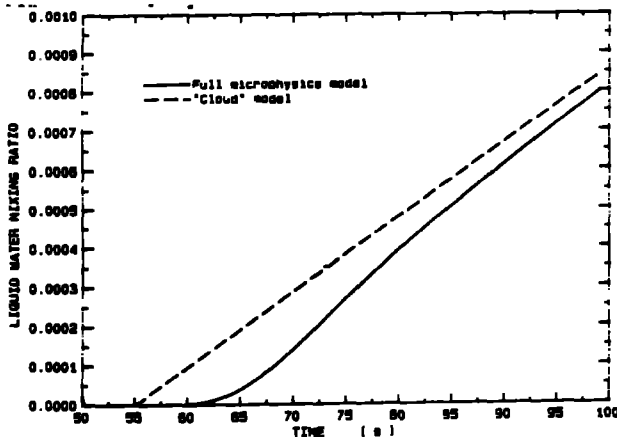


Figure 1. The liquid water mixing ratio is shown as a function of time, for a problem in which a constant updraft of 10 m/s was assumed. The solid line refers to a full microphysical model calculation in which an initial aerosol loading of 100 particles/cm³ was assumed. The dashed line refers to a calculation in which the cloud model assumption, $S_R \leq 1$, was assumed. Both calculations were initialized at a height of 1000 m with $T=288^\circ\text{K}$, $p=900$ mb and 75% relative humidity.

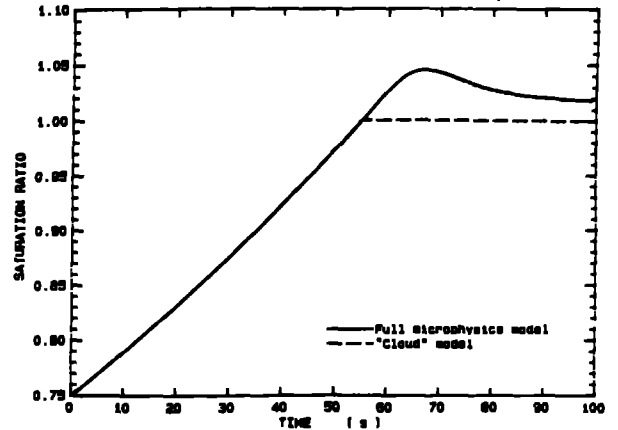


Figure 2. The saturation ratio is shown as a function of time for the two calculations described in Figure 1.

To calculate the saturation ratio, we first compute the decrease in drop and aerosol concentration due to dilution of the parcel by upward motion and spreading of the smoke plume:

$$\frac{\partial f_d}{\partial t} = \frac{-f_d}{r_{w,dil}}$$

$$\frac{\partial q_{AP,d}}{\partial t} = \frac{-q_{AP,d}}{r_{w,dil}} \quad (1)$$

$$\frac{\partial f_{AP_s}}{\partial t} = \frac{-f_{AP_s}}{r_{s,dil}}$$

$$\frac{\partial q_{AP,s}}{\partial t} = \frac{-q_{AP,s}}{r_{s,dil}}$$

where $r_{s,dil}$ and $r_{w,dil}$ are chosen to assure consistency with the cloud model:

$$r_{s,dil}^{-1} = (\rho_{s,c}^t - \rho_{s,c}^{t+\delta t}) / (\rho_{s,c}^t \cdot \delta t) \quad (2)$$

$$r_{w,dil}^{-1} = (\rho_{v,c}^t + \rho_{l,c}^t - \rho_{v,c}^{t+\delta t} - \rho_{l,c}^{t+\delta t}) / ((\rho_{v,c}^t + \rho_{l,c}^t) \cdot \delta t)$$

The liquid water in the microphysics model is computed according to

$$\rho_l = \int f_d(a) \cdot \frac{4}{3} \pi (a^3 - r_n^3) \rho_w da \quad (3)$$

$$+ \int f_{AP,s}(a) \frac{4}{3} \pi (a^3 - r_n^3) \rho_w da$$

where r_n is the dry particle radius and ρ_w is the density of water. The vapor density in the microphysics model can be computed from the difference between the total water (liquid plus vapor) in the cloud model and the liquid water density in the microphysics model:

$$\rho_0 = \rho_{l,c} + \rho_{v,c} - \rho_l \quad (4)$$

Next, these drop and aerosol distributions are used to estimate the change in liquid water due to condensation processes, following the method suggested by Hall (1980). Thus, an average value for S_R over the time step δt is found, consistent with the growth of drops and aerosols over that time step. In this procedure, the temperature is that predicted by the cloud model. The final liquid water content is then determined by summation over the drop and aerosol distribution (e.g. Eq. 3), and the microphysics model vapor density is found by difference (e.g. Eq. 4).

This method for coupling can be tested by first computing drop and aerosol distributions in a fully coupled parcel model. This constitutes an estimate for the "exact" distributions and values of the saturation ratio. Following this, the parcel model is run as a "cloud" model. In this computation drop growth and size distribution is ignored and ρ_l is computed by the assumption $S_R \leq 1$ at all times. A history tape of the variables required by the microphysics trajectory model is produced. Finally, the microphysics trajectory model is run, using this history tape as input. For the case shown in figure 1, we calculate a maximum supersaturation of 4.5% with the fully coupled model. The trajectory model predicts a significantly lower saturation of only 3.5%. This represents an extreme test, however, for studies of the nucleation of smoke over fires, since the particle loading is only 100 particles/cm³.

If the particle loading is increased to 10⁶ particles/cm³ and an updraft velocity of 50 m/s is specified, the calculated maximum supersaturation from the fully coupled model is 0.99% while that from the trajectory model is 0.89%. Thus the trajectory model is expected to give adequate results for the simulation of nucleation scavenging above fires, where copious numbers of particles are expected. At the meeting, we will present results from the trajectory model which are consistent with the dynamics, water concentration, and smoke distribution from a full three-dimensional hydrodynamic model calculation of a fire and its smoke plume.

3. MICROPHYSICS MODEL PARAMETER STUDIES

We have used our parcel model to test the dependence of the maximum predicted supersaturation on the assumed aerosol properties and updraft velocity. For these simulations, we assumed a fixed updraft velocity, rather than computing the updraft as is normally assumed in parcel models (see, for example, Flossman et al., 1985). We note that our model essentially reproduces the results of Flossman et al. (1985), for the same conditions considered by them. Here, our calculations with fixed updraft velocity simulate a situation in which the updraft is primarily forced by an external heat source such as that provided by a fire (Penner, et al., 1986). For simplicity, we also assume that the entrainment of background aerosol and drops is negligible.

The following calculations were initialized at an altitude of 1000 m, with a temperature of 288.16°K and pressure of 900 mb. The relative humidity was 75 percent. Figure 3 shows the computed saturation ratio for two cases of updraft velocity. For these calculations, the aerosol was assumed to be insoluble, but wettable. With this assumption, the aerosol is activated once the environmental saturation ratio becomes larger than the Kelvin term in the aerosol growth equation for a given particle radius. The aerosol size distribution was taken to be lognormal with a geometric standard deviation of 2.0. The saturation ratios for two different lognormal size distributions are shown in the figure. In each case the parcel was initialized with 10⁶ particles/cm³. Particle concentrations of this magnitude are much larger than those assumed under background conditions, but may be typical of the concentrations expected above a fire (Penner and Porch, 1986). For an updraft velocity of 10 m/s, the parcel becomes supersaturated after about 55 seconds or at an altitude of 1550 m. For updrafts of 50 m/s as expected above a large-scale, intense fire (Penner et al., 1986), the parcel reaches the same altitude and becomes supersaturated after only 10 seconds. For the same aerosol distribution, the higher updraft increases the maximum supersaturation by almost a factor of two. Thus the maximum supersaturation increases from 0.5% to 1.0% for a lognormal aerosol distribution with mode radius equal to 0.1 μ m and from 0.9% to 1.7% for a distribution with a mode radius of 0.05 μ m. Larger saturation ratios are reached with the smaller aerosol size distribution because fewer large particles are available to act as condensation nuclei when the parcel first becomes supersaturated. In the cases with updraft equal to 50 m/s, all particles with radii greater than 0.10 μ m and greater than 0.06 μ m are activated to drops for the size distributions with mode radii of 0.1 and 0.05 μ m, respectively. For an updraft velocity of 10 m/s, the corresponding activation radii are 0.20 μ m and 0.11 μ m, respectively.

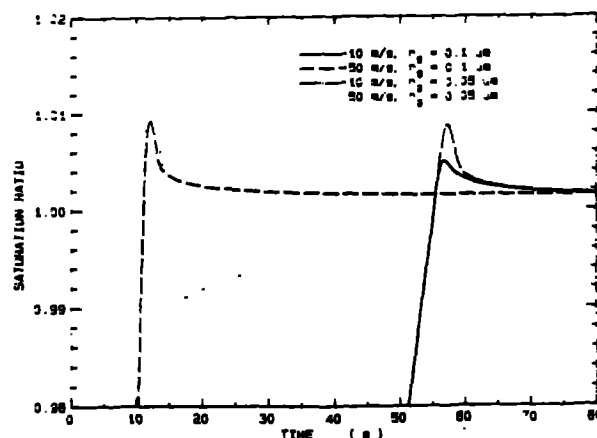


Figure 3. The saturation ratio is shown as a function of time for two vertical updraft velocities and two aerosol size distributions. The aerosol was assumed to be insoluble, but wettable, and the total number concentration was initialized to 10⁶ particles/cm³.

The effect of the total number of particles on saturation ratio is shown in figure 4. As the total number of aerosol particles decreases, the maximum supersaturation increases. In the case of a 10 m/s updraft velocity, for a mode radius of 0.1 μm , the maximum supersaturation increases from 0.5% with $10^6/\text{cm}^3$ to 1.5% with $10^4/\text{cm}^3$. Similarly, for a mode radius of 0.05 μm , the maximum supersaturation increases from 0.9% to 1.9%. With these higher supersaturations, all particles with radii larger than 0.07 μm and larger than 0.06 μm are incorporated into drops for the larger and smaller size distributions, respectively, even though the updraft velocity was only 10 m/s.

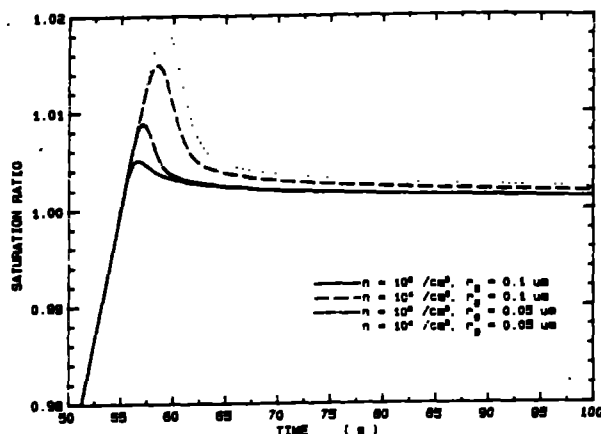


Figure 4. The saturation ratio is shown as a function of time for two initial number concentrations and two aerosol size distributions. The aerosol was assumed to be insoluble, but wettable, and the updraft velocity was 10 m/s.

Finally, figure 5 shows the effect of varying the aerosol composition. If the aerosol is fully soluble, only very small supersaturations are reached, because water is allowed to condense on the unactivated aerosol, reducing the tendency to supersaturation and swelling the particle radius so that the numbers of aerosol particles available to activate at any given saturation ratio is increased. For the fully soluble aerosol shown in figure 5, only particles with dry radii larger than 0.90 μm are activated.

4. CONCLUSIONS

We have developed a microphysical model that calculates the rate of condensation of water onto aerosols and their activation to drops. The model fully accounts for the effects of varying aerosol composition and size characteristics, at least for the assumption of spherical particles. At the present time, the model assumes that the composition of each aerosol size category is the same as the average aerosol composition, although we are developing a version of the model that does not include this constraint. Our results imply that substantial numbers of submicron aerosol particles may be incorporated into the capping clouds that are expected to form above large-scale fires. This should be accounted for in assessments of the effects of smoke on the atmosphere after a nuclear war.

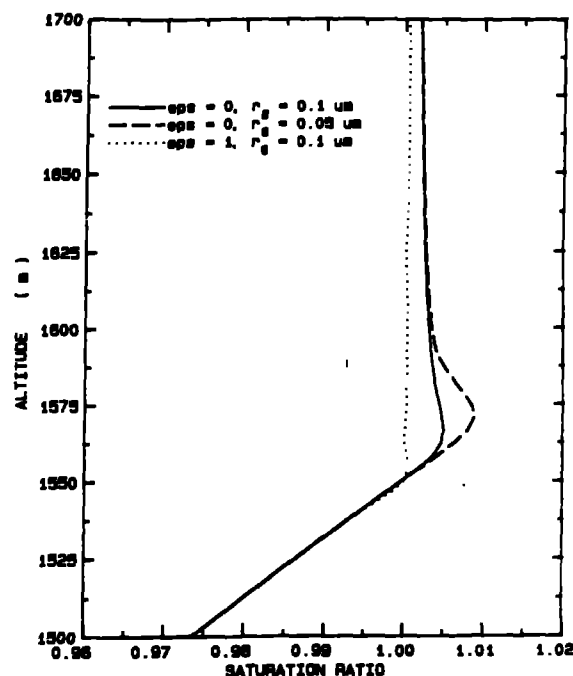


Figure 5. The saturation ratio is shown as a function of altitude for a fully soluble aerosol as well as an insoluble, but wettable aerosol. The initial aerosol number concentration was $10^6/\text{cm}^3$ and the updraft velocity was 10 m/s.

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